



Application No. 10/656,147
Appeal Brief

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF: :

Takashi MAEDA et al : GROUP ART UNIT: 1754

SERIAL NO.: 10/656,147 : EXAMINER: HENDRICKSON, S.

FILED: SEPTEMBER 8, 2003

FOR: PROCESS FOR PRODUCING MESOPHASE PITCH BASED ACTIVE CARBON FIBER, MESOPHASE PITCH BASED ACTIVE CARBON FIBER AND ELECTRIC DOUBLE LAYER CAPACITOR

APPEAL BRIEF

COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

SIR:

Appellants submit this brief in response to the Final Rejection dated June 28, 2006.

REAL PARTY OF INTEREST

Honda Motor Company, Ltd. is the real party of interest in the present application.

RELATED APPEALS AND INTERFERENCES

To the best of the Appellants' knowledge, there are no appeals or interferences which will directly affect or be directly affected by, or have a bearing on, the Board's decision in this appeal.

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STATUS OF CLAIMS

Original Claims 1-9 have been canceled. Claims 10-22 are pending.

STATUS OF AMENDMENTS

All amendments and remarks filed in this case have been entered and considered.

SUMMARY OF CLAIMED SUBJECT MATTER

Claim 10 is directed to a method of preparing an electric double layer capacitor by preparing carbon fibers by (i) carbonizing a mesophase pitch based infusibilized fiber at 600° to 900° C, (ii) activating the thus obtained carbon fiber in the presence of alkali, thereby obtaining a meso-phase pitch based active carbon fiber having a BET specific surface area of 30 to 1000 m²/g whose pores consist essentially of micropores having an average pore radius of 0.2 to 1 nm; forming positive and negative electrode material by formulating a mixture of said activated carbon fibers, conductive carbon black and a binder and applying the mixture to a solid conductive metal support; positioning the positive and negative electrodes so formed in non-contacting relationship in a container with an electrolyte; and subjecting the positive and negative electrodes to a charge/discharge treatment in which the capacitor is charged at constant current density at an increasing voltage until the voltage exceeds 2.5 V up to 3.5 V, thereby forming an electric double layer at the interfaces of the electrodes of the capacitor and the electrolyte and thereafter discharging the capacitor at a constant current density.

A concise explanation of the subject matter of Claim 10 is found on page 11 of the specification.

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Independent Claim 11 is directed to a method of increasing the charge/discharge capacities of activated carbon fiber for an electric double layer capacitor, by (i) carbonizing a mesophase pitch based infusibilized fiber at 600° to 900° C, (ii) activating the thus obtained carbon fiber in the presence of alkali, thereby obtaining a meso-phase pitch based active carbon fiber; and (iii) immersing the thus obtained carbon fiber in an electrolyte in which a current is applied at constant current density and at an increasing voltage until the voltage exceeds 2.5 V up to 3.5 V to the mesophase pitch based active carbon fiber with the result that an electric double layer is formed at an interface of the mesophase pitch based active carbon fiber and the electrolyte to thereby effect charging, and thereafter discharging the capacitor at a constant current density.

A discussion of the aspects of Claim 11 can be found on pages 25 to 29 of the specification.

GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL

Whether Claims 10 – 22 are unpatentable over Maeda et al, U.S. Patent 6,118,650 in view of Anani et al, U.S. Patent 5,439,756, Neat et al, U.S. Patent 5,030,523, and an alleged admission by the Applicants.

ARGUMENT

Claim 10

Claim 10 is directed to a method of preparing an electric double layer capacitor. Appellants have noted the comments that have been made concerning what is known in the

prior art, but respectfully submit that the Examiner has misstated what is known and what is unknown in the prior art. The three cited references disclose a method of making a claimed capacitor. Appellants have indeed stated on page 33, lines 8-11 of the specification that methods of preparing electrodes of capacitors are known and are not particularly limited and that such conventional methods can be used in the present invention without modification. However, the Examiner has improperly “carried over” this statement to the charge/discharge treatment step of the process embodiments of the present claims by which the electrode, immersed in an electrolyte, is subjected to the charge/discharge treatment at constant current density at increasing voltage until the voltage exceeds 2.5 V up to 3.5 V. (This treatment of the present invention is not to be confused with the repeated charge/discharge cycles that a capacitor is subjected to over the period of its use.)

Appellants have noted the Examiner’s comments concerning what is known in the prior art, but respectfully submits that he has not properly stated what is known and what is unknown in the prior art. The cited Maeda et al patent discloses a method of making a capacitor. Appellants have indeed stated on page 33, lines 8-11 of the specification that methods of preparing the electrodes of capacitors are known and that they are not particularly limited and that such conventional methods can be used in the present invention without modification. However, the Examiner has “carried over” this statement over to the charge/discharge treatment step of the process embodiments of the present claims by which the electrode, immersed in an electrolyte, is subjected to the charge/discharge treatment at constant current density at increasing voltage until the voltage exceeds 2.5 V up to 3.5 V. (This treatment of the present invention is not to be confused with the repeated charge/discharge cycles that a

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capacitor is subjected to over the period of its use.)

The specification states on page 30 that it has been believed that an active carbon fiber of large specific surface area in which pores having a certain specific size which permit infiltration of an electrolyte are distributed in large amounts through the fibers is desirable for use as an electrode material in electric double layer capacitors. In the present invention the mesophase based active carbon fiber on the basis of the charge/discharge capacity of the active fiber per unit weight is highly associated with its the specific surface area and pore distribution. However, in the process of achieving the present invention, it has been found that the mesophase pitch based active carbon fibers obtained in the activation step, when having a small specific surface area as measured by the BET method and when containing micropores having been regarded as unsuitable to the formation of an electric double layers in large amounts and thus having a small average pore radius that is calculated from the t-plot of a nitrogen absorption isotherm, exhibits a marked effect of increased charge/discharge capacity in the charge/discharge treatment step. If this charge/discharge treatment step is repeated again, the resulting increase in the effect of the invention is not as high as obtained from the initial treatment, but substantially high charge/discharge capacities can be maintained thereafter under regular charge/discharge conditions.

The Examiner maintains the combination of the Maeda et al patent with the Anani et al and Neat et al patents and appellants' submissions. However, Maeda et al in columns 7 to 12 teaches the features of the newly added claims, but differs in not teaching the details of the use and construction of the battery/cell, however, the specification on page 33 states that these details are old and known.

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The Neat et al patent, in column 3, appears to disclose the charging of a cell to 3.25 volts, and a carbon containing material. Anani et al discloses in column 4 a charging scheme which can be varied according to the system in what appears to be the same electrode synthesis and an active carbon material.

As to the matter concerning Claims 12 and 13, column 4 of Maeda et al indicates that most values of the patent are overlapped by the reference. Although most of the examples disclose an area greater than 1000, this value, nevertheless, can be optimized by the values described in column 4 by varying the time of activation. Moreover, with respect to Claims 17, 21 and 22, the use of a non-aqueous electrolyte is an obvious expedient to do chemistry of organic materials, while immersing an electrode is an obvious expedient to permit current to flow and reagent to contact each other in normal operation.

As to the matter of Anani et al, the same describes that upon full charge of the cell, the electrical storage device is allowed to equilibrate before the discharge step is applied. The operating voltage window is typically between 0.8 V to 1.7 V. Moreover, Anani et al describes that the battery capacity of the electrical storage device is about 25 mAh, as measured by a slow discharge of 1 mA current.

As to the Neat et al patent, the same discloses an electrochemical cell that has an active anode material of an alkali metal such as lithium. A solid alkali metal ion conducting electrolyte is also disclosed. Further, Neat discloses that the cell is charged at a constant current at the C/10 rate to an upper voltage limit of 3.25 V. The cell took a large degree of charge and is able to deliver about 60 % of the theoretical discharge capacity on the second discharge cycle. Subsequent cycling under the same conditions indicates a minimal loss in discharge

capacity over the next twenty cycles.

However, the charging schemes disclosed in Anani et al and Neat et al are merely characteristic evaluations of a secondary electrochemical cell. It is not obvious to apply the schemes of the references in order to prepare an electrical double layer capacitor or to the preparation of an activated carbon fiber. It must be remembered that an evaluation of characteristics must be distinguished from preparation. Accordingly, it would not have been obvious to one of skill in the art, at the time the present invention was made, to use the charging scheme disclosed in order to improve the charge/discharge capacities of the cell in order to improve upon the charge/discharge capacities for preparing activated carbon fiber and a condenser. Furthermore, the specific treatment disclosed which requires the immersion of the carbon fiber in an electrolyte and repeating the charge/discharge cycle of Claim 11, the charge/discharge of a secondary electrochemical cell can be conducted in the highly mixed state of a carbon fiber/electrolyte.

It is not clear to applicants precisely what the mechanism is by which the charge/discharge treatment of the invention results in the improvement of the present invention. Since it does not appear that the charge/discharge cycle of the treatment results in a significant change in the specific surface area and the average pore radius of the active carbon fibers, it can be presumed that there is some change with respect to the structure and/or internal-surface properties of pores that have small radii as a result of the treatment. These pores of small radii may contribute along with the pores having larger radii so to result in an electrode that has a marked increase in charge and discharge capacities. (see pages 31 and 32 of the text) The specific treatment of the present invention is not shown or suggested by the Maeda et al patent.

Claim 11

Claim 11 is directed to a method of increasing the charge/discharge capacities of activated carbon fiber for an electric double layer capacitor. In simplified form, the method comprises three stages:

- 1) carbonizing a fiber by heating the fiber;
- 2) activating the carbonized fiber; and
- 3) placing the activated fiber in an electrolyte and applying the current while increasing the voltage such that an electric double layer is formed at the interface of the activated fiber and the electrolyte, and then effecting a discharge.

The Examiner has rejected Claim 11 as being obvious over the combination of Maeda, Anani, Neat and an alleged admission by the Applicants at page 33 of the specification. The Examiner is incorrect for the following reasons:

At the outset, Maeda does not describe or suggest stage 3) of the inventive process of Claim 11 (i.e., placing the fiber in an electrolyte and applying current while increasing voltage to form an electric double layer and then effecting a discharge). Rather, the fibers of Maeda involve only a carbonization step (please see, for example, Maeda at column 6, lines 40-41, and 51-53) and an activation step (please see, for example Maeda at column 7, lines 51-55).

The Examiner then wrongly alleges that the charge/discharge stage of Claim 11 is admitted as being old and known in the art, by the Applicants, at page 33 of the specification (please see the Official Action, page 2, which describes: "The Examiner takes Official Notice that these specific techniques are old and known, as is the discharge/charge under constant

current.” As previously stated, the stage 3) of method Claim 11 involves a charge/ discharge of the fibers. Page 33, lines 8-11, of the specification describes: “In the present invention, the method of preparing the electrode is not particularly limited, and any conventional electrode producing techniques can be used without modification.” The words “discharge/charge under constant current” are not described anywhere in the alleged admission. Further, Applicants submit even a “creative” reading of Page 33, lines 8-11, would not produce that a “discharge/charge under constant current,” as part of a process for producing improved fibers (i.e., the third process stage of Claim 11), was admitted as being old and known. Applicants note that the charge/discharge treatment of the present invention is not to be confused with the repeated charge/discharge cycles that a capacitor is subjected to over the period of its use. Accordingly, the alleged admission, with regard to the charge/discharge under constant current, is no admission at all, and the combination of Maeda and the alleged admission do not suggest or describe all of the limitations of present Claim 11.

Regarding Neat: there is no suggestion or motivation to combine Neat with Maeda. Further, even if there were a suggestion to combine Neat and Maeda, there is no expectation of success, except with the hindsight of the Applicants’ invention.

Neat, at column 3, lines 20-31, describes the manufacture and “proving” (i.e., showing that the cell works) of an electrochemical cell. Applicants note the cell has Manganese (IV) oxide (MnO_2) as the primary cathode material (please see column 1, lines 3-4, of Neat) and an anode material of lithium, sodium, potassium, or a combination thereof (please see column 1, lines 47-53, of Neat). Neat describes that the cathode can contain graphite (please see column 2, line 8). Neat then describes constructing an electrochemical cell from the anode and cathode

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and “proving” the cell (i.e., charging and discharging the cell) at column 3, lines 20-32.

Applicants submit there is no suggestion or motivation to combine the Neat with Maeda. Neat describes making a using an electrochemical cell which may contain graphite. The graphite is not activated and is not a fiber. Where is the motivation to take the charging and discharging of the electrochemical cell and use the charging and discharging as a stage in a process for creating an improved carbon fiber? Applicants submit the Examiner has not supplied the motivation to combine references and that any combination of Neat with Maeda is therefore improper.

Additionally, even if there were motivation to combine Neat with Maeda, where is the expectation of success (i.e., a process that results in a fiber with improved properties)? Applicants submit there is no expectation of success (except with the hindsight of the Applicants’ invention), and the combination of Neat and Maeda is improper for this reason.

Finally, Anani describes that upon full charge of the cell, the electrical storage device is allowed to equilibrate before the discharge step is applied. The operating voltage window is typically between 0.8 V to 1.7 V. Moreover, Anani describes that the battery capacity of the electrical storage device is about 25 mAh, as measured by a slow discharge of 1 mA current.

However, the charging schemes disclosed in Anani are merely characteristic evaluations of a secondary electrochemical cell. It is not obvious to apply the schemes of the references in order to prepare an activated carbon fiber. It must be remembered that an evaluation of characteristics must be distinguished from preparation. Accordingly, it would not have been obvious to one of skill in the art, at the time the present invention was made, to use the charging scheme disclosed in order to improve the charge/discharge capacities of the cell in

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order to improve upon the charge/discharge capacities for preparing activated carbon fiber and a condenser. Accordingly, the combination of Anani and Maeda does not describe or suggest the process of instant Claim 11, and instant Claim 11 is therefore not obvious in view of the combination of Anani and Maeda.

Because Claim 11 is not obvious in view of the cited references, Applicants submit Claims 12-13 and 18-21, which depend from Claim 11, are also non-obvious.

Based on the above made arguments, Appellants believe the Examiner's decision to continue rejection of the claims is erroneous and should be REVERSED.

Respectfully submitted,

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APPENDIX I (CLAIMS ON APPEAL)

Claim 10. A method of preparing an electric double layer capacitor, comprising preparing carbon fibers by:

(i) carbonizing a mesophase pitch based infusibilized fiber at 600° to 900° C,

(ii) activating the thus obtained carbon fiber in the presence of alkali, thereby obtaining a meso-phase pitch based active carbon fiber having a BET specific surface area of 30 to 1000 m²/g whose pores consist essentially of micropores having an average pore radius of 0.2 to 1 nm;

forming positive and negative electrode material by formulating a mixture of said activated carbon fibers, conductive carbon black and a binder and applying the mixture to a solid conductive metal support;

positioning the positive and negative electrodes so formed in non-contacting relationship in a container with an electrolyte; and subjecting the positive and negative electrodes to a charge/discharge treatment in which the capacitor is charged at constant current density at an increasing voltage until the voltage exceeds 2.5 V up to 3.5 V, thereby forming an electric double layer at the interfaces of the electrodes of the capacitor and the electrolyte and thereafter discharging the capacitor at a constant current density.

Claim 11. A method of increasing the charge/discharge capacities of activated carbon fiber for an electric double layer capacitor, comprising:

(i) carbonizing a mesophase pitch based infusibilized fiber at 600° to 900° C,

(ii) activating the thus obtained carbon fiber in the presence of alkali, thereby obtaining

a meso-phase pitch based active carbon fiber; and

(iii) immersing the thus obtained carbon fiber in an electrolyte in which a current is applied at constant current density and at an increasing voltage until the voltage exceeds 2.5 V up to 3.5 V to the mesophase pitch based active carbon fiber with the result that an electric double layer is formed at an interface of the mesophase pitch based active carbon fiber and the electrolyte to thereby effect charging, and thereafter discharging the capacitor at a constant current density.

Claim 12. The method as claimed in Claim 11, wherein the activated carbon fiber has a BET specific surface area of 30 to 1200 m²/g.

Claim 13. The method as claimed in Claim 11, wherein the activated carbon fiber has an average pore radius of 0.2 to 1 nm.

Claim 14. The method as claimed in Claim 10, wherein the carbon fiber is milled to a particle size in terms of average particles in the range of 5 to 50 μ m prior to activation step (II).

Claim 15. The method as claimed in Claim 10, wherein said activation of the carbon fiber occurs in the presence of alkali metal compound in a weight ratio of alkali metal compound to carbon fiber of 0.5 to 5.

Claim 16. The method as claimed in Claim 10, wherein said activation is conducted in an inert gas at a temperature of 500 to 900° C.

Claim 17. The method as claimed in Claim 10, wherein said electrolyte in the charge/discharge treatment is an organic nonaqueous electrolyte.

Claim 18. The method as claimed in Claim 11, wherein the carbon fiber is milled to a particle size in terms of average particles in the range of 5 to 50 μm prior to activation step (II).

Claim 19. The method as claimed in Claim 11, wherein said activation of the carbon fiber occurs in the presence of alkali metal compound in a weight ratio of alkali metal compound to carbon fiber of 0.5 to 5.

Claim 20. The method as claimed in Claim 11, wherein said activation is conducted in an inert gas at a temperature of 500 to 900° C.

Claim 21. The method as claimed in Claim 11, wherein said electrolyte in the charge/discharge treatment is an organic nonaqueous electrolyte.

Claim 22. The method as claimed in Claim 10, wherein the electrodes are immersed in the electrolyte of the container.

APPENDIX II (EVIDENCE)

No evidence has been relied upon in the record of the case pursuant to paragraphs 1.130, 1.131 or 1.132 of this title or any other evidence entered by the Examiner.

APPENDIX III
(RELATED PROCEEDINGS APPENDIX)

None.